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MEASUREMENT OF Pu CONTAMINATION AT THE 10-nCi/g LEVEL
IN 55-GALLON BARRELS OF SOLID WASTE
WITH A ^{252}Cf ASSAY SYSTEM

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ABSTRACT

A combination active/passive nondestructive assay (NDA) system is described for the measurement of a wide range of plutonium-contaminated solid waste. The waste packaged in 55-gal (208 liter) barrels could be of either high or low density with gamma-ray radiation from fission products as high as 1000 R/h. The passive NDA measurement technique consists of counting coincident and total neutrons with ^{3}He-filled proportional detectors. The active assay, which is made after the passive measurement, employs a cyclical irradiation by a ^{252}Cf neutron source followed by delayed neutron counting with the source transferred to a shielded storage container. Approximately 1000 s are used for the combination active/passive assay. The three standard deviation detectability limit is near the 10-nCi/g fiducial for the passive plutonium measurement and below the fiducial for the active measurement for barrels with a mass of 100 kg or more.

1. INTRODUCTION

The investigation of nondestructive assay (NDA) techniques for 55-gal (208 liter) barrels of solid waste was initiated as part of a nuclear safeguards program for reactor fuel reprocessing plants(1). The characteristics of the items to be assayed were left fairly general so the technique would be suitable for a wide range of waste categories. The waste materials considered included combustible waste such as rubber gloves or paper wipes, low-density waste such as air filters, high-density waste such as failed process equipment cut up to fit into 55-gal barrels, and waste generated during standard chemical analysis procedures. The radiation level from fission products was considered to be variable and potentially high enough to require remote handling(2). The chemical and isotopic compositions of the uranium or plutonium were assumed to be unknown.

Given the rather wide boundaries on the materials to be assayed, certain NDA techniques can be readily eliminated from consideration. Techniques employing passive gamma-ray and x-ray detection would not be practical because the large container size and high density would result in absorption of most of the signal, and interference from the fission product activity would mask the remaining signal(3). Techniques based on accelerators were eliminated because the practicality of routine waste measurement needs to be demonstrated. The remaining techniques are limited to those that employ gamma-ray insensitive gas-filled neutron detectors and active techniques that employ isotopic neutron sources. The techniques in this category include passive neutron counting with thermal neutron gas-filled proportional detectors(4-6) and three generic types of active assay systems; alpha-n neutron source irradiation with coincidence counting of induced fission neutrons(7), photo-neutron source interrogation with fast fission neutron counting(1,8,9), and ^{252}Cf neutron interrogation with delayed fission neutron counting(10). All the above techniques will handle gamma-ray radiation levels of up to about 1 R/h with no modifications, and the radiation levels can be extended essentially indefinitely by using lead shielding.

Passive neutron counting has been demonstrated to be practical for plutonium assay. A unit installed at the plutonium recovery area at the Los Alamos Scientific Laboratory (LASL) has routinely assayed waste materials in barrel-size containers for the past eight years(11). The detectability limit for the existing unit is about 1-g Pu. (The detectability limit is defined as the quantity of material that produces a response three standard deviations above background in a thousand-second assay.) Depending on the type of information needed and isotopes other than plutonium that might be present in the waste, passive neutron counting has certain limitations. Passive counting is not very sensitive to uranium because of the low neutron yields for the ^{235}U and ^{238}U isotopes(3). Passive neutron counting primarily measures the spontaneously fissioning isotopes (^{238}Pu, ^{240}Pu, ^{242}Pu, ^{242}Cm, and ^{244}Cm) by coincidence counting, and the total plutonium content is calculated from the isotopic composition. The total neutron count is a sensitive indicator of plutonium but inaccurate because of the wide variation in the count rate for different chemical forms (a factor of 25 between oxide and fluoride). Also, other isotopes such as ^{242}Cm and ^{244}Cm can be interpreted as plutonium.

The active assay techniques primarily measure the fissile isotopes (^{235}U, ^{239}Pu, and ^{241}Pu). The assay technique employing neutron interrogation with coincident fission neutron counting is similar to a passive instrument except a neutron source is used to induce fissions instead of passively counting spontaneous fissions(7). Because of the neutron background produced by the source, the technique is best suited for measuring large quantities of ^{235}U. The fissile detectability limit in barrel-size containers is expected to be greater than 10 g making the instrument unsuitable for low-level waste assay. The photo-neutron interrogation with energetic fission neutron detection has the potential for a fissile detectability limit at the 1- to 10-mg level. However, because the intrinsic detection efficiency for fast neutrons with gas-filled proportional counters is low, the instrument cannot be used effectively for passive assay. In addition, the detection technique is sensitive to the waste matrix material. Moderation, especially by hydrogenous material, rapidly reduces the neutron energy below the detection threshold resulting in an erroneously low assay. The system employing ^{252}Cf neutron interrogation with delayed neutron counting has the advantages of being operable as a passive system, a high sensitivity to fissile isotopes in the active mode, and a comparatively low sensitivity to matrix effects. Thus, as the most versatile NDA instrument for a wide range of solid waste categories, the combination of passive neutron counting and ^{252}Cf interrogation with delayed neutron counting was investigated through detailed computer calculations and prototype test measurements.

2. MEASUREMENT PRINCIPLES

2.1 Passive Assay

The passive assay measures the naturally occurring neutrons emitted by plutonium or other isotopes present in the waste. The neutron radiation arises from either spontaneous fission or from alpha-particle interactions with light elements. Spontaneous fission has been observed for thorium and heavier elements; however, for low-level waste measurements only the even isotopes of plutonium and curium make a significant spontaneous fission neutron contribution to the passive counting. An average of two or more neutrons are emitted simultaneously in the spontaneous fission process. The passive assay uses the coincidence of the detected neutrons to determine presence and quantity of spontaneously fissioning isotopes. The neutrons resulting from the alpha decay process are emitted singly. The neutron energy and emission rate varies drastically with the various light elements; for pure metallic pieces of the alpha-emitting isotopes only the spontaneous fission neutrons are available for detection.

Neutrons emanating from the container are moderated to near thermal energies by hydrogenous material surrounding the gas-filled detectors. Because the system is also operated in neutron interrogation mode, activated charcoal-lined ^{3}He-filled detectors must be used to avoid neutron damage during the irradiation by the ^{252}Cf source(12). The statistical variation in the time interval required to moderate the neutrons necessitates the use of coincidence gate times on the order of 100 μs. The gate time is long enough that accidental coincidences from the singly emitted alpha-n neutrons, background neutrons, or neutrons from different spontaneous fissions can be significant. Depending on the neutron emission rates, quite sophisticated electronics and data-reduction algorithms may be required to separate the "true" coincidences from the "accidental" coincidences. However, electronic units are commercially available that automatically account for the accidental coincidences at high count rates and for variable background rates(13).

The detectors can be operated in gamma-ray fields up to about 1 R/h. If the items to be assayed have radiation levels above 1 R/h, then lead shielding is installed between the waste barrel and the detectors. A lead shield approximately 5-cm thick is required to attenuate the more energetic fission product gamma rays by a factor of ten(14). Because lead has a very low neutron absorption cross section, the shield can be made as thick as necessary to reduce the gamma-ray radiation below 1 R/h at the detectors, but more neutron detectors would be required to surround both the shield and the item being assayed. The lead shield surrounding a 55-gal barrel designed to reduce a 100 R/h radiation level to 1 R/h would be 10-cm thick and have a mass of about 4000 kg. Cosmic-ray interactions with this quantity of lead can result in an appreciable neutron background rate, i.e., a background coincidence rate equivalent to about 9 g of ^{240}Pu at the Los Alamos altitude of 2200 m(15). At sea level the cosmic rays are attenuated by a factor of 4 and an overhead cover of concrete 1-m thick reduces the background another factor of 20. Thus, for a typical facility at sea level a background coincidence rate equivalent to 0.1 g ^{240}Pu can be expected from the lead shielding. In some cases it may be practical to operate the unit with more overhead shielding, such as in a tunnel, in which case the background from cosmic rays becomes negligible(16).

2.2 Active Assay

The active assay measures the fissile isotopes present in the waste by counting the delayed neutrons after the item has been irradiated by a ^{252}Cf neutron source. The assay unit consists of an isolated source storage position and a sample interrogation and counting chamber as shown in Fig. 1. The assay begins with the source in the storage position and the waste barrel in the assay chamber. The data accumulated with the neutron detector banks is then equivalent to a passive assay and both the total and coincident neutron rates are accumulated. The counting time for the passive portion of the assay is approximately five minutes (300 s). The active portion of the assay is cyclical consisting of irradiation by the ^{252}Cf source and delayed neutron counting with the source withdrawn. The irradiation time interval is about 16 s, the delayed neutron counting time interval is about 8 s, and the source transfer time is about 0.5 s in both directions resulting in a period of 25 s for one active assay cycle. The complete assay of passive counting and 28 active cycles requires 1000 s. A delayed neutron counting time shorter than the irradiation time is used to increase the assay precision by improving the delayed neutron signal to background ratio.

A high-speed stepping motor is used to transfer the source between the storage and irradiate positions. The unit shown in Fig. 1 has two irradiation positions to help make the interrogation uniform along the height of the barrel. An alternative scheme would translate the source vertically yielding a continuous scan during the irradiation portion of the assay.

The neutrons emitted from the californium source have a fission energy spectrum with an average energy of about 2.3 MeV. Moderating material surrounds the source at the interrogation position (not shown in Fig. 1) so that the barrel is irradiated with near thermal energy neutrons. The low-energy neutron irradiation enhances the response because of the increased fission cross section at thermal energies. The response can also be increased by using a more intense ^{252}Cf source. Practical trade offs between the desired detectability limit for a reasonable assay time and the quantity of radiation shielding needed for personnel safety result in a ^{252}Cf source size of about 2-5 mg or a maximum neutron rate of 10^{10}/s. The ^{252}Cf half-life is 2.65 years and the initial source strength is usually selected to provide about five years of service.

Delayed neutrons are emitted from some fission fragments after they have undergone one or more beta transitions and are characterized by their parent nuclei half-lives, which range from 0.2 to 55 s. The delayed neutron yield of ^{239}Pu fission is about 0.6%(17). Neither delayed neutrons nor prompt neutrons from fission can be observed during the irradiation of the sample because the ^{252}Cf source saturates the response of the detectors. Thus, the source must be transferred to the shielded storage position during the delayed neutron counting.

The neutron detectors completely surround the interrogation and counting chamber shown in Fig. 1. With the detectors in a "4π" geometry the detection efficiency is maximized and the neutron response is uniform over the sample volume. The item is rotated during the assay to improve the uniformity of the neutron irradiation.

3. PROTOTYPE TEST MEASUREMENTS

The prototype test measurements concentrated on investigating the active assay technique of neutron irradiation and delayed neutron counting for 55-gal barrels. The passive neutron assay of barrels has been investigated at LASL and reported(11,15,18). Advances in the passive method will result primarily from replacing $^{10}BF_3$-filled detectors with more efficient ^{3}He-filled detectors and increasing external shielding to reduce background neutrons.

A test bed was assembled by constructing a barrel-size assay chamber(14) for an existing small sample assay unit(10). The test bed provided the data needed to estimate the expected performance of a fully engineered system such as shown in Fig. 1. In addition the test bed served as a reference point for checking the accuracy of the LASL neutron Monte Carlo transport code (MCN)(19) by comparing the results with the observed performance of the test bed unit.

The test bed provided the data for the background neutron rates expected for barrel-size active assay systems and experience for reducing their contribution(20). The background rates, which affect both the active and passive measurements, arise from ambient room neutrons, cosmic-ray interactions in the lead shielding, and neutrons from the ^{252}Cf source at its storage position. The room background rate was 300 counts/s with only a cadmium sheet covering the outside surface of the detector banks. After installing a 20-cm-thick polyethylene shield the room background rate was reduced to 4 counts/s. In strictly passive assay units with no lead liner the room background is the only background. The 15-cm-thick, 4800-kg lead shield with a 0.9-m-thick concrete overburden at the Los Alamos altitude of 2200 m produced a background of 14 counts/s. A thick overburden of earth or concrete is not unreasonable for a facility routinely processing materials with high gamma-ray radiation levels. Thus, the need for cosmic-ray shielding to reduce background rates is consistent with the usual requirements to limit radiation levels outside the facility. The test bed unit had an observed background count rate of 10 counts/s from the 0.2-mg ^{252}Cf source at its storage position. The transfer tube length between the storage position and the assay chamber was 1.37 m with one 3-deg-bend midway to prevent a line-of-sight neutron path. Because the source background results primarily from neutrons streaming down the transfer tube, additional distance and bends will be effective in further reducing the source background contribution.

The Monte Carlo calculations were made in two steps(21). First the number of fissions induced in the barrel per source neutron was estimated. The fission rate was tallied by position in the barrel to check uniformity. Because of the single point irradiation used on the test bed the fission rate varied by nearly a factor of four over the barrel volume. In the second step of the Monte Carlo calculation the delayed neutron detection efficiency was obtained. The average detection efficiency was 10% but varied by a factor of three from the center to the ends of the barrel because the detectors were placed only on the sides and end losses were enhanced by the 15-cm-thick lead shield. The agreement between the Monte Carlo calculations and the observed performance for the test bed was quite good, typically within ±15%. The slight differences generally resulted from "simplifications" used in the calculation geometry.

Although the test bed was constructed to test the feasibility of low-level measurements (<1 g fissile), a request was made to use the instrument to assay highly enriched uranium (93%, ^{235}U) intermixed with furnace parts and reduction residues. The material was packaged in 30-gal (114 liter) barrels and the uranium content had been estimated using a "by difference" technique(22). In addition to the test bed a Segmented Gamma Scanner (SGS)(23) was used for passive counting of the 185-keV gamma rays from ^{235}U. Both techniques used a single standard containing 46.4-g ^{235}U intermixed with paper in a 30-gal barrel to approximate the barrels of waste. The SGS used a gamma-ray transmission measurement to correct for the matrix differences between the standard and the waste barrels; however, no matrix corrections were made for the neutron-based test bed measurements.

Table I shows the results of two NDA assay techniques along with their one-standard-deviation statistical precision. For the 10 barrels assayed by both the SGS and the test bed, the total ^{235}U inventory was 177.6 g and 165.9 g, respectively, a reasonable agreement for this type of material. (Two barrels were not assayed by the neutron technique, because they were too heavy to transport to the test bed.) The higher count rate for the neutron technique gave better precision than the gamma-ray measurement. For the standard, the neutron NDA test bed count rate was 7.4 counts/s/g ^{235}U; whereas, the SGS count rate was 1.8 counts/s/g ^{235}U. However, because the waste barrels were more dense than the standard, the SGS count rate was reduced by about an order of magnitude.

4. EXPECTED PERFORMANCE

The detection limit using neutron irradiation and delayed counting with the test bed unit is about 67 mg ^{239}Pu and the detection limit for the passive assay unit at the LASL plutonium facility(11) is about 60 mg ^{240}Pu by coincidence counting. Figure 1 shows a system designed to optimize the detection limit for both the active and passive neutron measurements. The improvements in the design include a more efficient neutron detection scheme using a "4π" geometry. The tubes are filled with ^{3}He and thereby give a higher efficiency than the BF_3 models previously used in the passive barrel-size systems. Monte Carlo calculations yield a

TABLE I

^{235}U CONTENT OF TWELVE 30-GAL BARRELS CONTAINING FURNACE PARTS AND REDUCTION RESIDUES MEASURED BY THE SGS AND THE NEUTRON TEST BED

Drum No.	Net Weight (kg)	Estimated ^{235}U (g)	Measured ^{235}U[a]	
			SGS	Delayed Neutron
70	92	65	59.1 ± 2.2	...
79	31	9	53.8 ± 1.4	37.50 ± 0.10
80	23	6	1.1 ± 1.0	1.239 ± 0.016
81	56	16	4.8 ± 1.4	3.699 ± 0.030
82	47	12	2.1 ± 1.0	2.314 ± 0.026
83	126	61	55.9 ± 1.4	...
84	69	36	14.8 ± 0.8	19.72 ± 0.06
85	68	35	19.2 ± 1.0	18.89 ± 0.06
86	68	35	20.8 ± 0.9	24.84 ± 0.08
87	69	30	21.6 ± 1.0	21.67 ± 0.07
88	69	28	21.3 ± 0.9	20.46 ± 0.07
89	39	33	18.8 ± 1.0	15.54 ± 0.06

[a]Errors indicate statistical precision only (1-σ level).

neutron detection efficiency in either the active or passive mode of 35%(24). The presence of neutron absorbers in the matrix material or the inclusion of a cadmium liner in the assay chamber to reduce the matrix material sensitivity may limit the detection efficiency to about 25%. A larger 2-mg ^{252}Cf source improves the detectability limit. The source would yield nearly 5 x 10^9 n/s and be about 10 times more intense than the source in the test bed unit. The shielding on the storage unit (0.76-m thick from source to surface) limits the personnel radiation exposure to about 1 mR/h.

In quoting the detectability limits in the active and passive modes, a conservative approach is taken in estimating the background rates at a waste processing facility. The rates are extrapolated from the active assay test bed and from the passive 30-gal assay unit installed at the LASL plutonium facility TA-55(11). For estimating purposes the plutonium isotopic composition is assumed to be 80% ^{239}Pu and 20% ^{240}Pu and the plutonium is in the oxide form (PuO_2). For the instrument shown in Fig. 1, the five-minute passive portion of the assay yields a detectability limit using coincidence counts of 30-mg Pu without the lead shield and 200-mg Pu with the 15-cm-thick lead shield. An additional overburden of 1-2 m thickness would eliminate the background from the lead. The passive neutron totals count rate has a detection limit of 25-mg Pu with the lead shield. The substitution of plutonium fluoride (PuF_4) for the oxide would adversely affect the coincidence detectability limit and improve the total count detectability limit. The active portion of the assay measures the induced delayed neutron rate and accounts for about 700 s of the 1000-s assay. The active portion of the assay has a detectability limit of 4-mg Pu and the chemical form does not affect the limit. For barrels with 100 kg of waste (density = 0.5 g/cm^3) 1 mg Pu corresponds to approximately 1-nCi/g contamination. Thus, the active technique is capable of sorting waste at the 10-nCi/g fiducial and the passive techniques are close to the

10-nCi/g fiducial. Combining the coincidence and integral passive counting data with the active assay results yields the fertile and fissile plutonium components as well as the probable chemical composition.

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CAPTIONS

Figure 1. A combination active/passive ^{252}Cf assay system for 55-gal barrels.